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Domain Formation and Molecular Alignment in Nematic Liquid Crystal Mixtures with Positive Dielectric Anisotropy

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The relative effectiveness of electric and magnetic fields for producing molecular alignment and consequent instabilities has been investigated in nematic liquid crystal mixtures of MBBA and MBCA with positive dielectric anisotropy in moderately thick samples. Domain formation and its stability has been related to the magnitudes of positive dielectric anisotropy. Grid patterns have been observed in mixtures with dielectric anisotropy between +0.5 and +1.2. The variation of threshold voltage with dielectric anisotropy for domain formation and dielectric alignment in moderately thick and thin samples has been reported. It is found that the maximum values of positive dielectric anisotropy below which electrohydrodynamic instability is likely to exist in thin and moderately thick samples differ.

INTRODUCTION

The effect of electric fields on nematic liquid crystals with positive dielectric anisotropy and consequent instabilities like domain formation and dielectric alignment have been reported by several authors.[1–13] The transition from domain formation to dielectric alignment in nematics with positive dielectric anisotropy has been related to the magnitude of the dielectric anisotropy and to the ratio of dielectric

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constants. de Jeu[10] reported that the domains are predicted for a value of $\epsilon_{\parallel}/\epsilon_{\perp} < 1.05$. Domain formation and their stability have been related to the relative effectiveness of conduction and dielectric torques by Carr *et al.*[6] Theoretically Gruler[5] investigated the threshold behaviour of Williams domains and dielectric alignment at different signs and magnitudes of the dielectric anisotropy. Penz[14] reported a two dimensional treatment for the less positive $\Delta\epsilon$ nematics, the calculated threshold which involves dielectric and conductivity effects agrees with the Freederiksz expression above a dielectric constant ratio of $\epsilon_{\parallel}/\epsilon_{\perp} = 1.2$. Electrohydrodynamic (EHD) instability in materials of positive dielectric anistropy has been reported by Barnik *et al.*[11] The dependence of the threshold voltage on dielectric anisotropy under constancy of the other parameters have been investigated, and the experimental data is in agreement with the two dimensional theory of S. A. Pikin[15-16] in which boundary conditions are taken into account for Helfrich model. Zenginoglou *et al.*[12] investigated numerically in two dimensions, the ability of homogeneously aligned nematics with positive dielectric anisotropy to exhibit Williams domains and showed that domain formation can be extended in positive $\Delta\epsilon$ nematics by the application of stabilizing magnetic field.

The theoretical and experimental results show that the type of instability in positive $\Delta\epsilon$ nematics primarily depends on the relative magnitudes of conductivity and dielectric anisotropies in addition to some experimental conditions like type of alignment (homogeneous or homeotropic) and sample thickness. Most of the previous investigations regarding EHD instability in positive $\Delta\epsilon$ nematics was confined to thin samples except Carr's work on thick samples. The previous results suggest a value of $\Delta\epsilon = +0.5$ in thin samples below which EHD instability is likely to exist. But Carr's work suggests a value of $\Delta\epsilon = +1.0$ in thick samples. The present work is aimed at investigating EHD instability in moderately thick and thin samples of MBBA (*p*-methoxy benzylidene *p'*-*n*-butyl aniline and MBCA (*p*-methoxy benzylidene *p'*-Cyano aniline) mixtures with positive dielectric anisotropy in the presence of electric and magnetic fields.

EXPERIMENTAL

Dielectric constant measurements are made at a frequency of the order of applied electric field since this frequency is of primary importance in the orientation of the molecular axes and consequent instabilities. Due to experimental limitations, dielectric constant measurements have been carried out at 100 kHz using crystal controlled oscillator. We believe that 100 kHz frequency is well below the dispersion region for MBBA–MBCA mixtures since the concentration of MBCA at which the dielectric anisotropy of the mixture changes sign nearly agrees with the values reported earlier[17] at 1 kHz on the similar type of mixtures.

The dielectric constant measurement at 1 MHz[13,18] have been used to study changes in the molecular alignment. The rf field of frequency 1 MHz is below the dispersion region and it is only a probing field to elicit information regarding the state of molecular ordering.

The MBBA and MBCA samples have been prepared in our laboratory using standard procedures. MBBA has been subjected to fractional distillation under reduced pressure and MBCA has been recrystallized twice before use. The concentration of MBCA in MBBA used in this work is very low, so the viscosity of the mixture would not be much different from that of MBBA. *Ac* conductivities of the mixtures are of the order of 10^{-10} ohm^{-1} cm^{-1}. The conductivity ratio $\sigma_{\parallel}/\sigma_{\perp}$ is 1.3 for MBBA, measured at 80 Hz. This shows slight increase as the MBCA concentration increases.

A uniform and moderate sample thickness of 250 μm is used for the present work. Sample thickness of 40 μm is employed for the experimental investigations in thin samples. Homogeneous alignment in thin samples is obtained by rubbing technique. The threshold voltages in thin samples are measured by using photo multiplier and the optical observations are made with a microscope.

RESULTS AND DISCUSSION

(A) Dielectric constants of MBBA - MBCA mixtures

The dielectric constants $\epsilon_{\parallel}$ and $\epsilon_{\perp}$ have been measured for ten samples of varying concentration of MBCA in MBBA at 100 kHz and 32°C, sample thickness = 250 μm. A magnetic field of 5.7 KG is used to align the molecular axes parallel and perpendicular to the electric field.

Figure 1 represents a plot of dielectric anisotropy $\Delta\epsilon = \epsilon_{\parallel} - \epsilon_{\perp}$ of the mixtures as a function of concentration of MBCA in MBBA by weight percentage at 100 kHz. The points are experimental data and the solid line is calculated assuming additivity relation (19). The dielectric anisotropies of MBBA and MBCA are -0.41 and $+17.0$ at 100 kHz and 32°C respectively. The resulting values of $\Delta\epsilon$ calculated

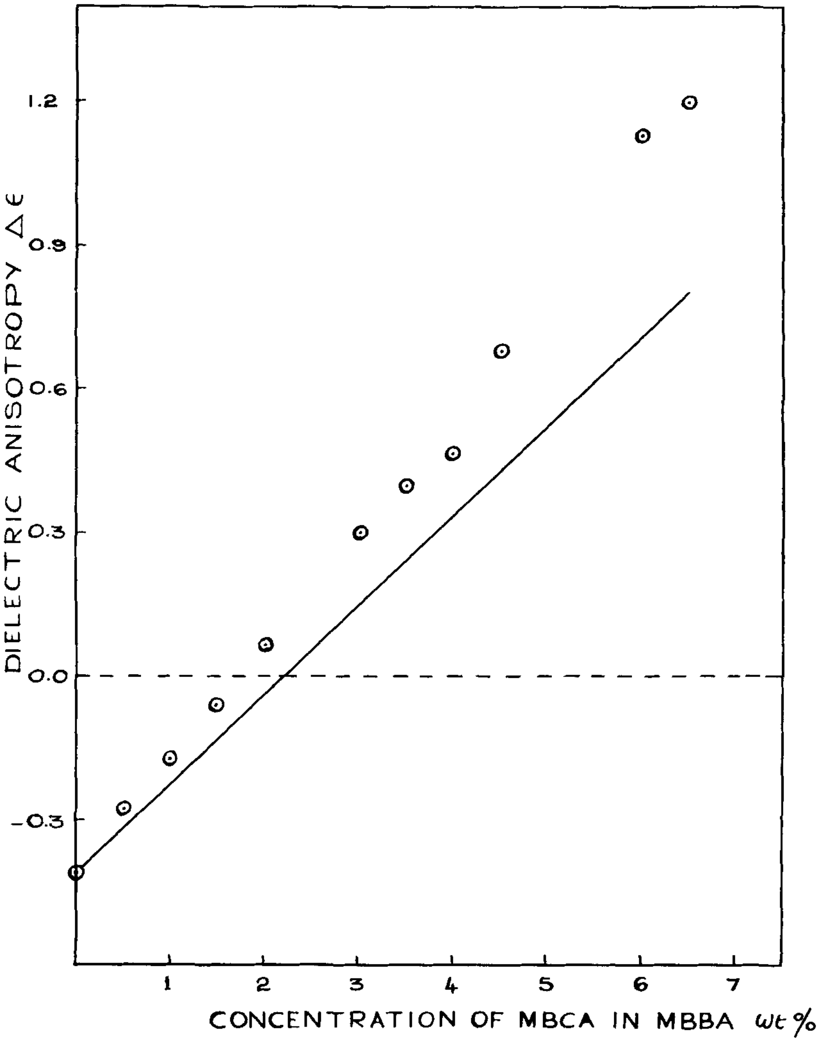

FIGURE 1 Dielectric anisotropy, $\Delta\epsilon$ of MBBA-MBCA mixtures as a function of weight percentage of MBCA in MBBA at 100 kHz. Temperature = 32 °C and H = 5.7 KG.

from the additivity relation differ with the experimental data, this may be a temperature effect.[19] The point of dielectric isotropy occurs nearly at 1.8% by weight of MBCA in MBBA.

(B) Molecular alignment and domain formation

This section deals with the electric and magnetic field effects on the molecular alignment in relation with the instabilities on MBBA—

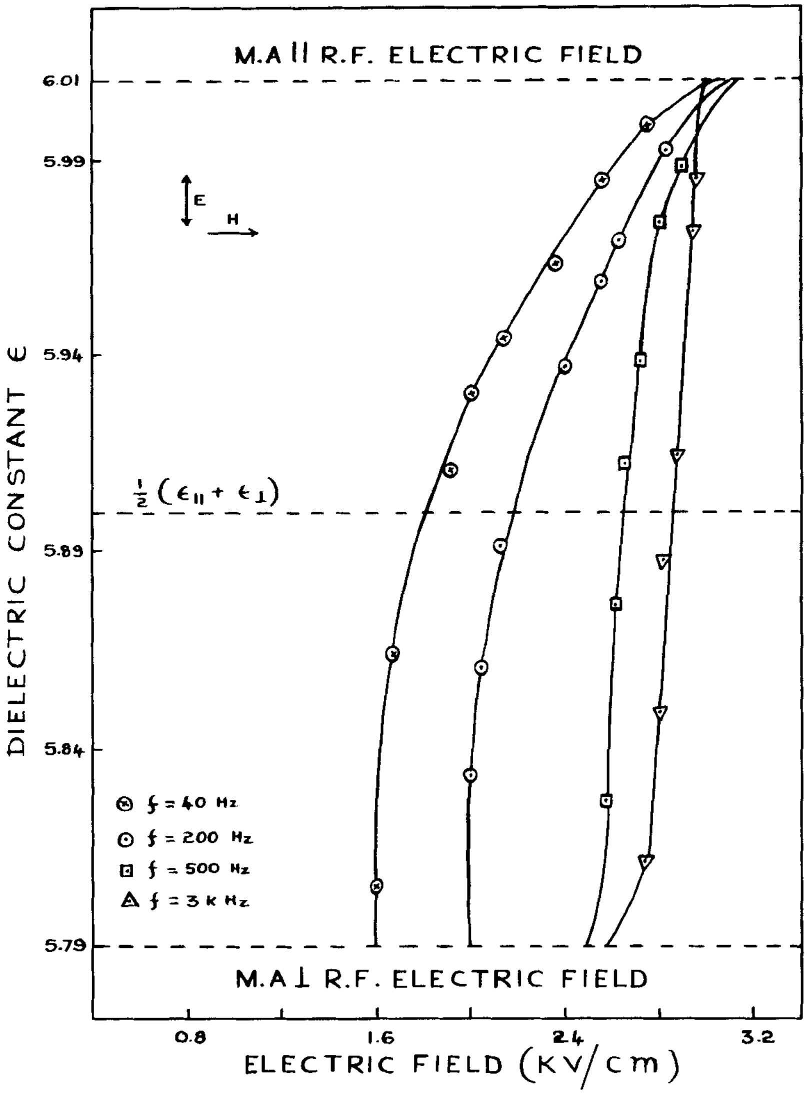

FIGURE 2 Dielectric constant, ϵ of 3% MBBA–MBCA mixture at 1 MHz as a function of externally applied electric field of various frequencies. $H \perp E$, $H = 5.7$ KG and Temperature = 32 °C. M.A. = Molecular Axes.

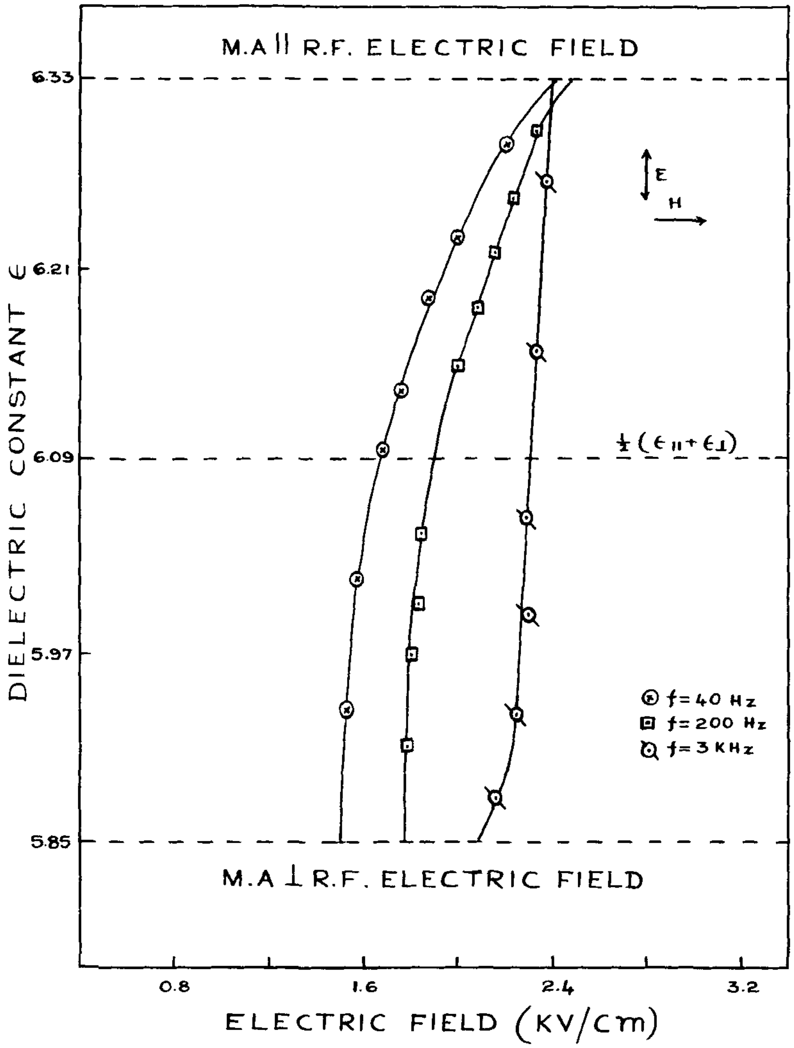

FIGURE 3 Dielectric constant, ϵ of 4% MBBA-MBCA mixture at 1 MHz as a function of externally applied electric field of various frequencies. $H \perp E$, $H = 5.7$ KG and Temperature = 32° C. M.A. = Molecular Axes.

MBCA mixtures with positive dielectric anisotropy. The nature of instability has been related to the magnitude of positive dielectric anisotropy measured at 100 kHz. Figure 2 shows the frequency dependence of the molecular alignment in the presence of 5.7 KG magnetic field applied perpendicular to the external electric field for a concentration of 3% by weight of MBCA in MBBA with $\Delta\epsilon = +0.3$ at 100 kHz.

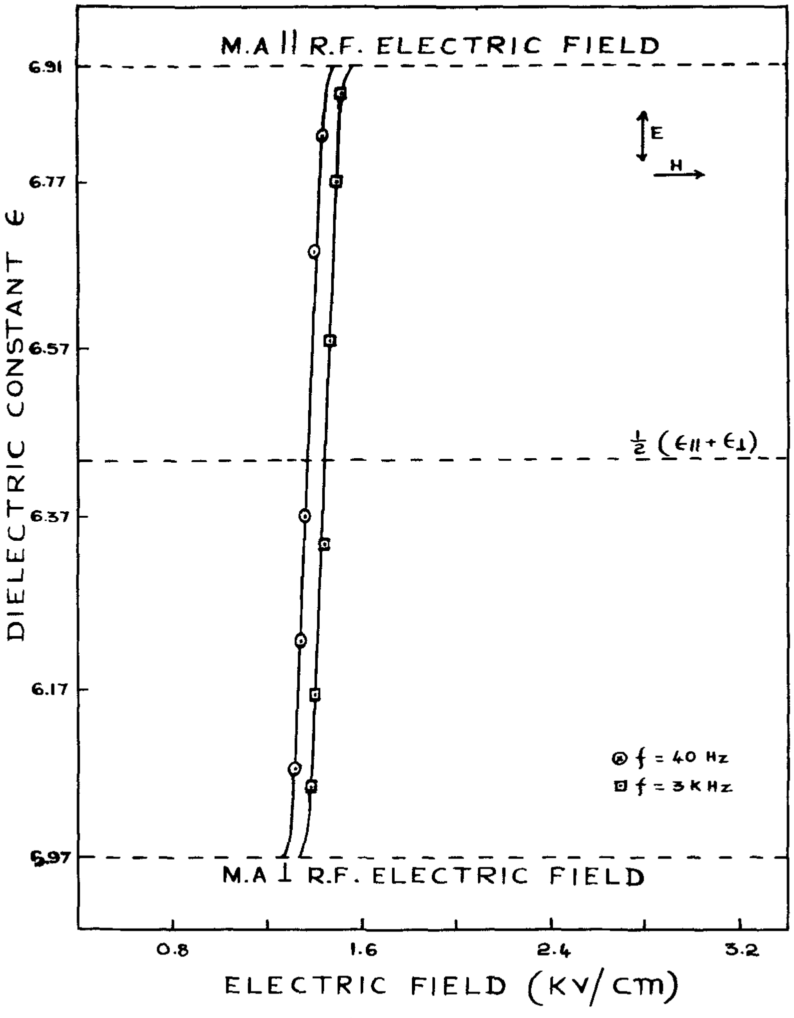

FIGURE 4 Dielectric constant, ϵ of 6.5% MBBA–MBCA mixture at 1 MHz as a function of externally applied electric field of various frequencies. $H \perp E$, $H = 5.7$ KG and Temperature = 32 °C. M.A. = Molecular Axes.

The results indicate that there exist a critical frequency $f_c = 600$ Hz separating conduction and dielectric regimes.[6] The threshold voltage for domain formation is frequency dependent in the conduction regime.[13] The torques due to the conductivity and dielectric anisotropies aid each other in causing parallel molecular alignment in the conduction regime. But in the dielectric regime, reorientation of the molecular axes from perpendicular alignment to the parallel alignment involves torque due to the dielectric anisotropy only.

It has been reported earlier[6] that the domain formation in positive $\Delta\epsilon$ nematics can be related to the relative effectiveness of torques due to the conductivity and dielectric anisotropies. Figure 2 represents that the conduction torque is larger than the dielectric torque in the conduction regime. Stable domains and weak scattering are observed.

The results of 4% and 6.5% concentrations with dielectric anisotropies +0.46 and +1.2 at 100 kHz are shown in Figures 3 and 4. For comparison, Figures 2, 3 and 4 are plotted with the same scale of electric field intensity. Figure 3 illustrates that the conduction and dielectric torques are nearly equal in the conduction regime, unstable domains are observed. Figure 4 shows that the dielectric torque is larger than the conduction torque in the conduction regime, only pure reorientation has been observed.

(C) Variation of threshold Voltage with $\Delta\epsilon$

The variation of threshold voltage with dielectric anisotropy has been investigated by Gruler[5] and Barnik *et al.*[11] Figure 5 illustrates the variation of the threshold voltage for domain formation (V_{th}) and dielectric alignment (V_F) with dielectric anisotropy measured at 100 kHz for MBBA–MBCA mixtures, sample thickness = 250 μm.

The line 1 represents the variation of V_{th} with dielectric anisotropy in the $H \perp E$ condition. The curve 2 illustrates the variation of V_F with positive $\Delta\epsilon$ in the $H \perp E$ condition. The curve 3 shows the variation of V_F with negative $\Delta\epsilon$ in the $H \parallel E$ configuration.

It is clear from Figure 5 that V_F decreases with the increasing values of the dielectric anisotropy. But V_{th} decreases gradually as $\Delta\epsilon$ changes from -0.41 to $+1.2$. As the positive values of $\Delta\epsilon$ increases, the threshold difference between V_{th} and V_F decreases and finally reaches a near value at $+1.2$. According to the optical observations, the positive $\Delta\epsilon$ values are divided as follows:

(a) $\Delta\epsilon < +0.35$	Stable domains and DSM
(b) $+0.35 < \Delta\epsilon < +0.5$	Unstable domains
(c) $+0.5 < \Delta\epsilon < +1.2$	Grid pattern
(d) $\Delta\epsilon \geq +1.2$	Pure reorientation

The results indicate that the EHD instability exists up to a value of $\Delta\epsilon = +1.2$, and domain formation below $+0.5$. This agrees nearly with the results of Carr[6] in thick samples. The dielectric constant ratio $\epsilon_{\parallel}/\epsilon_{\perp} = 1.2$ at 100 kHz for 6.5% concentration agrees with the theoretical value of Penz.[14]

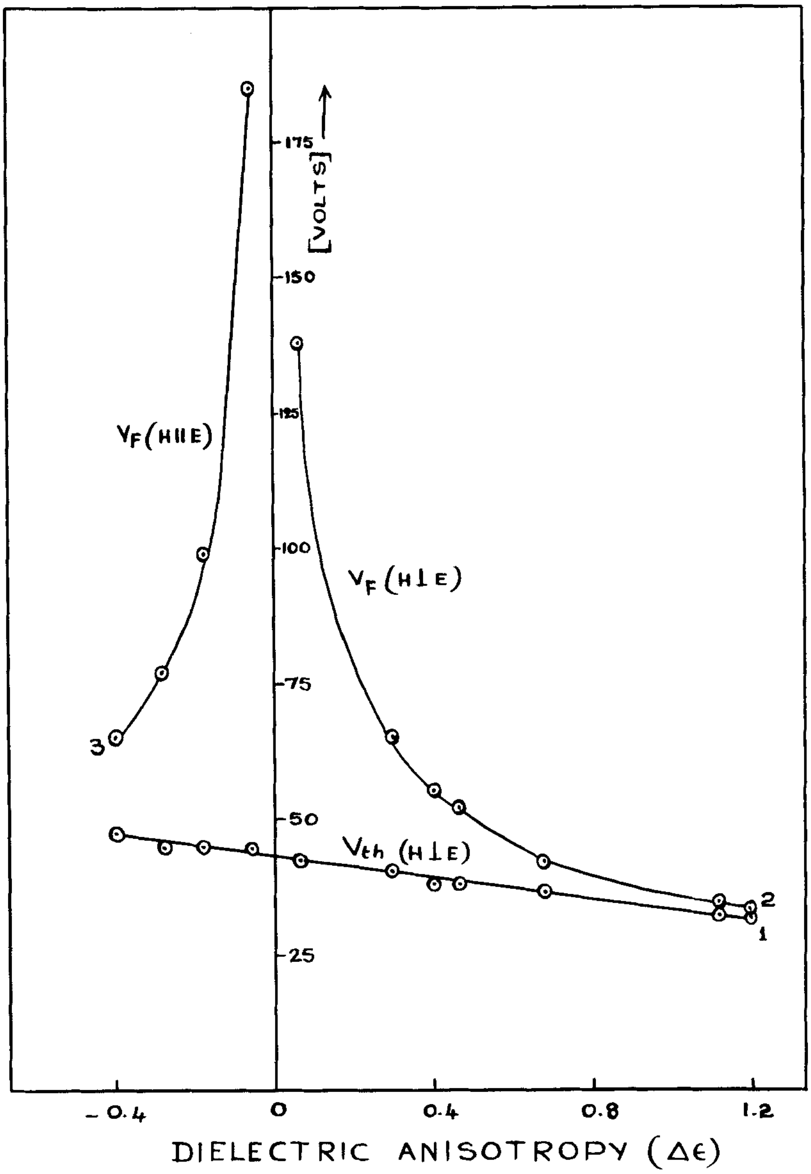

FIGURE 5 The threshold voltage for domain formation (V_{th}) and dielectric alignment (V_F) in MBBA–MBCA mixtures as a function of the dielectric anisotropy $\Delta\epsilon$ at 100 kHz. H = 5.7 KG, Temperature = 32° C and sample thickness = 250 μm.

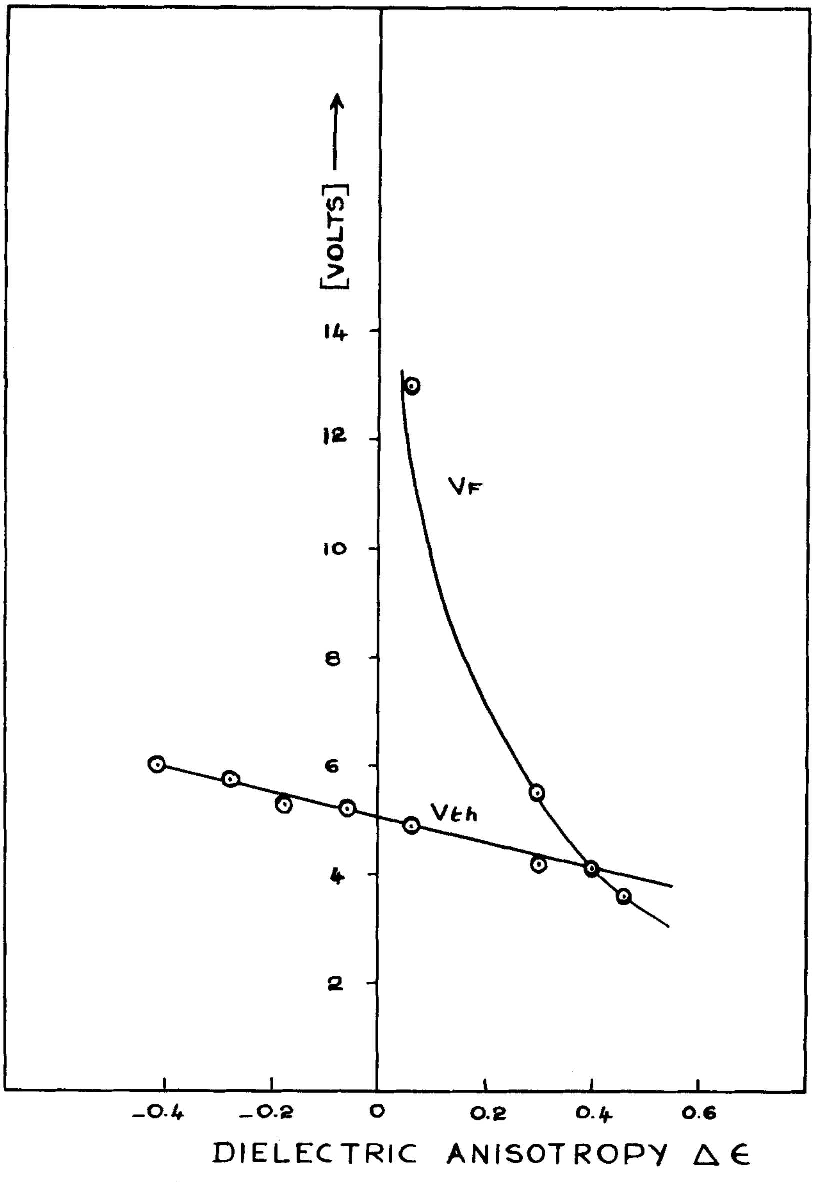

FIGURE 6 The threshold voltage for domain formation (V_{th}) and dielectric alignment (V_F) in MBBA-MBCA mixtures as a function of the dielectric anisotropy $\Delta\epsilon$ at 100 kHz. $H = 0$, Temperature, $= 32°C$ and sample thickness $= 40\ \mu m$.

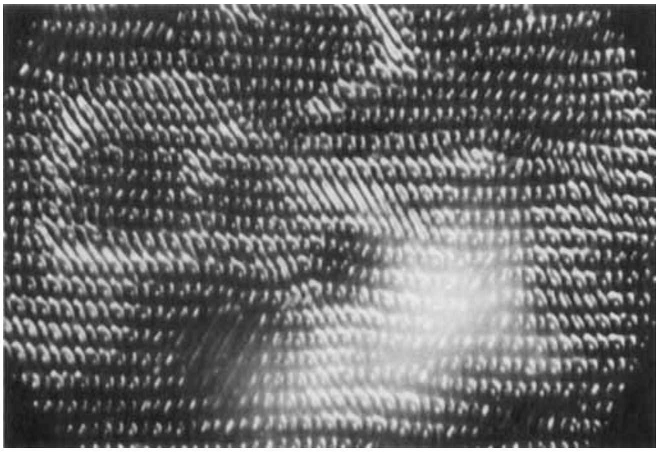

FIGURE 7 Grid Pattern. $H \perp E$, $H = 5.7$ KG, $\Delta\epsilon = +0.68$ at 100 kHz, Temperature $= 32°$C.

EHD instability studies are also extended to thin homogenously orientated samples (40 μm) with the same mixtures. Figure 6 shows the variation of V_{th} and V_F with $\Delta\epsilon$. This suggests a value of $\Delta\epsilon = +0.4$ above which EHD instability does not exist. At the low values of $\Delta\epsilon = +0.1$, stable domains and turbulence are observed. Between $+0.1$ and $+0.4$, unstable domains and reorientation exist. At and above the values of $+0.4$, there is only reorientation.

The above results indicate that domain formation in moderately thick and thin samples nearly exist up to a value of positive $\Delta\epsilon = +0.5$, which agrees with the previous observations.[6] EHD instability and domain formation in thin samples nearly exist up to the same value of positive dielectric anisotropy. But in the case of moderately thick samples, the EHD instability extends from $+0.5$ to $+1.2$ in the form of grid pattern as shown in Figure 7.

It is evident from the above results that the conductivity instability has been suppressed in thin samples to the lower values of positive $\Delta\epsilon$ as compared to moderately thick samples. Recent theoretical work of Smith *et al.*[20] also suggests that the conductivity instability can be suppressed in thin samples.

CONCLUSIONS

The nature of $\Delta\epsilon$ variation with the concentration of MBCA in MBBA is linear and agrees with the previous observations. Relative study of molecular alignment and EHD instability or domain formation in positive $\Delta\epsilon$ nematic mixtures show that the type of instability can be characterized by the relative effectiveness of conduction and dielectric torques. The maximum values of positive $\Delta\epsilon$ below which EHD instability is likely to exist in thin and moderately thick samples differ. This is due to the elimination of conductivity instability in thin samples to lower positive $\Delta\epsilon$ values as compared with thick samples.

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